

Numerical test of the damping time of layer-by-layer growth on stochastic models

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We perform Monte Carlo simulations on stochastic models such as the Wolf-Villain (WV) model and the Family model in a modified version to measure mean separation ℓ between islands in submonolayer regime and damping time \tilde{t} of layer-by-layer growth oscillations on one dimension. The stochastic models are modified, allowing diffusion within interval r upon deposited. It is found numerically that the mean separation and the damping time depend on the diffusion interval r , leading to that the damping time is related to the mean separation as $\tilde{t} \sim \ell^{4/3}$ for the WV model and $\tilde{t} \sim \ell^2$ for the Family model. The numerical results are in excellent agreement with recent theoretical predictions.

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Recently problems of surface growth by molecular beam epitaxy (MBE) have been attractive in statistical physics [1]. From the point of view of statistical physics, it is intriguing how various stochastic processes involving many atoms such as shot noise, diffusion, and nucleation, are described in terms of scaling structures. These cooperative phenomena depend crucially on the ratio between the diffusion constant D and the deposition rate F . The deposition rate is defined as number of atoms landing on surface per unit area and unit time. Atoms deposited on surface diffuse until they meet one another to form dimers which then grow into islands of monoatomic height. The mean distance of nucleation events, corresponding to the mean separation between islands, is determined by the ratio between the diffusion constant D and the deposition rate F as [2-8]

$$\ell \sim (D/F)^\gamma. \quad (1)$$

The exponent γ depends on diffusion process of adatoms and islands shape. It is also a function of the critical island size i^* , defined by the size $i^* + 1$ of the smallest island which is stable enough that it never decays before capturing next adatom. When only adatoms can move and desorption can be neglected, the exponent γ can be predicted using the kinetic theory as [6]

$$\gamma = \frac{i^*}{2i^* + d + d_f}. \quad (2)$$

However, recently the formula was corrected for the case of $d = 1$ and $i^* \geq 2$ as [9]

$$\gamma = \frac{i^*}{2i^* + 3}. \quad (3)$$

As number of adatoms increases in submonolayer, islands grow in their sizes and coalesce into forming bigger islands, and eventually cover a monolayer. If adatoms are allowed to diffuse to stable sites such as kink sites or step

edges before other adatoms getting deposited and if interlayer diffusion is not inhibited by the Ehrlich-Schwoebel barrier [10,11], smooth surface with a minimum of defects is grown. In this case, surface exhibits a layer-by-layer growth [12]. As islands evolve, the density of atomic steps oscillates, which is a hallmark of layer-by-layer growth. The oscillating behavior can be monitored through the RHEED intensity in laboratory, which is useful for being aware of film thickness easily. Layer-by-layer growth is important for the fabrication of microelectronic devices. It is known that the oscillation persists perfectly when system size is smaller than the layer-coherence length $\tilde{\ell}$ such that for two sites within the coherence length $\tilde{\ell}$, they grows coherently, and beyond which they are out of phase [13]. The coherence length is related to the separation between islands as $\tilde{\ell} \sim \ell^{4/(4-d)}$. The coherence length is much larger than other length scales such as the islands separation ℓ or the characteristic length $\ell_0 \sim (D/F)^{1/(2+d)}$, where ℓ_0 is formed by a dimensional combination of D and F , meaning that if system size is smaller than ℓ_0 , it can accommodate at most one island. When system size is larger than $\tilde{\ell}$, the oscillation of step density is damped due to various types of fluctuations. It is recently found [13] that there exists a characteristic time \tilde{t} , beyond which the damped oscillation disappears. The characteristic time is scaled as

$$\tilde{t} \sim (D/F)^\delta, \quad (4)$$

meaning the critical time for the transition from layer-by-layer growth to kinetic roughening growth.

In the kinetic roughening growth, when number of adatoms is conserved, surface growth may be described by the equation, [11,13,14,15]

$$\partial_t h = -\nabla \cdot \mathbf{j} + \eta, \quad (5)$$

where h means surface height, \mathbf{j} is adatom current, and $\eta(\mathbf{x}, t)$ denotes shot noise satisfying $\langle \eta(\mathbf{x}, t) \rangle = 0$ and having the correlation,

$$\langle \eta(\mathbf{x}, t)\eta(\mathbf{x}', t') \rangle \sim \delta^d(\mathbf{x} - \mathbf{x}')\delta(t - t'), \quad (6)$$

with substrate dimension d . When adatom current is driven by the gradient of surface curvature and square of surface tilt with coefficients K and λ [11,13,14,15],

$$\mathbf{j} = \nabla[K\nabla^2 h - \lambda(\nabla h)^2]. \quad (7)$$

The continuum equation for surface growth is written as

$$\partial_t h = -K\nabla^4 h + \lambda\nabla^2(\nabla h)^2 + \eta(x, t), \quad (8)$$

which, called the conserved Kardar-Parisi-Zhang (cKPZ) equation, has been discussed in the context of MBE growth. For the cKPZ universality class, it was derived that the exponent δ for the damping time is related to γ for the islands-separation as [13]

$$\frac{\delta}{\gamma} = \frac{4d}{4-d}. \quad (9)$$

The derivation was based on dimensional analysis on the continuum equation. The formula was checked numerically in one dimension performing the coarse graining Monte Carlo simulations [13].

For the surface growth driven by downhill current with coefficient ν ,

$$\mathbf{j} = -\nu\nabla h, \quad (10)$$

the continuum equation is written as

$$\partial_t h = \nu\nabla^2 h + \eta(x, t), \quad (11)$$

which is called the Edwards-Wilkinson (EW) equation [16]. In this case, adatoms tend to move in downward direction on surface, which is realized by exchanging mobile atom with adatom at step edge as observed in homo epitaxial growth on Ir(111) [17]. In this case, the exponent δ for the damping time is related to γ as [13]

$$\frac{\delta}{\gamma} = \frac{2d}{2-d}, \quad (12)$$

which was also derived based on dimensional analysis on the continuum equation, Eq.(11). However, successful numerical confirmation of Eq.(12) has not been reported yet. Our preliminary numerical result for MBE growth with downhill current does not fit well to Eq.(12), requesting numerical check of the formulae, Eqs.(9,12) on stochastic models belonging to the cKPZ and EW universalities, respectively. In this letter, we perform Monte Carlo simulations on the stochastic models, the Wolf-Villain (WV) model [18] and the Family model [19] with some modifications in their dynamic rules to check Eqs.(9,12).

Let us first consider the modified WV model, of which the dynamic rule is defined as follows. First, a site, say

i -th site, is selected randomly on a one-dimensional flat substrate with system size L . Then we consider a subset of the system, composed of $2r + 1$ sites, the randomly selected site, and its $2r$ neighboring sites on its right and left sides, respectively, within distance r . Among the $2r + 1$ sites, surface is advanced at the site offering largest binding, that is, most occupied neighbors. If sites offering largest binding exists more than one, then a site closest to the i -th site among them is taken and its height is increased by one. The case of $r = 1$ is reduced to the original version of the WV model. If the distance r is regarded as diffusion length, then the diffusion constant would be related to the distance as $D \sim r^2$. We measure the density of islands in submonolayer regime and the surface fluctuation width W^2 [20],

$$W^2(L, t) = \left\langle \frac{1}{L} \sum_i (h_i - \bar{h})^2 \right\rangle, \quad (13)$$

with $\bar{h} = \sum_i h_i / L$ as varying the diffusion length r . Note that the density of islands ρ is related to the mean separation ℓ as $\rho \sim \ell^{-1}$ in one dimension, and the damped oscillation in the step density occurs in the surface fluctuation width in the same way. In our simulations, we used the noise reduction method with the noise reduction parameter $m = 5$ to get better data. However, we did not vary the parameter m in any case, so that the noise reduction parameter does not play a role of control parameter as considered in Refs. 21-22. As shown in Fig.1, the density of islands well behaves as $\rho \sim r^{-0.94}$ for large values of r , suggesting that the exponent $\gamma \approx 0.47$. The numerical result is close to the theoretical result $\rho \sim r^{-1}$, which is obtained as follows. If we divide the system into small cells with equal size $2r + 1$, and a particle is deposited on a cell randomly, then one island would be formed on each cell, and the separation of islands in the system varies as $\ell \sim r$ for large r . Thus $\rho \sim r^{-1}$. However, the cell boundary is virtual and could be overlapped to each other in our simulations, but the correction due to the fluctuation of boundaries would be of higher order. Thus the density of islands would behave as $\rho \sim r^{-1}$ up to leading order, and $\gamma \approx 1/2$. We also count number of nucleation event by ignoring monomers, and the result obtained is the same as Fig.1. Next, the surface fluctuation width W^2 exhibits a damped oscillation as shown in Fig.2(a). The data for different diffusion lengths are well collapsed, even for small values of r , when time is rescaled as t/\tilde{t} with $\tilde{t} \sim r^{4/3}$ as shown in Fig.2(b). The rescaled time suggests $\delta = 2/3$. Therefore, the ratio of the two exponents γ and δ is obtained as $\delta/\gamma \approx 4/3$, which is consistent with the theoretical prediction, Eq.(9) for $d = 1$.

Next, we consider the modified Family model. In this case, we choose a site randomly, say i -th site, on one dimension substrate with system size L , and consider a subset of $2r + 1$ sites as before. Among the $2r + 1$ sites, surface is advanced at lowest site. If sites with

the lowest height exist more than one, a site closest to the i -th site is taken. The case of $r = 1$ is reduced to the original version of the Family model. We vary the distance r , and examine the density of islands and the damped oscillating behavior of the surface fluctuation width. The density of islands in submonolayer regime behaves as the case of the WV model. For the surface fluctuation width, the data are well collapsed even for small values of r when time is rescaled as t/r^2 as shown in Fig.3(a), implying that the damping time behaves as $\tilde{t} \sim r^2$. Thus the ratio between the exponents δ and γ is obtained as $\delta/\gamma \approx 2$ for the Family model, which is consistent with the theoretical prediction, Eq.(12). We also check the case involving a hump on the way to the lowest site in the subset. In Fig.3(a), we considered the case that adatom can run over the hump, and stay on the lowest site within the interval. In Fig.3(b), adatom cannot run over hump, and is allowed to move only in a descending way or on the same height. We cannot find any difference between the two in the collapsing behavior, implying the assumption of the dephasing length \tilde{l} , within which the layer difference is at most one, is valid. Therefore, we conclude that the theoretical prediction is correct for the stochastic models, the WV model and the Family model, belonging to the cKPZ and EW universalities. However, the relevance of the theoretical prediction to the MBE growth with downhill current is to be further investigated.

In summary, we have performed Monte Carlo simulations on the Wolf-Villain model and the Family model involving the diffusion length r on one dimension. We measure the density of islands ρ in submonolayer regime and the damping time \tilde{t} for the oscillation of layer-by-layer growth as varying the diffusion length. We obtained numerically that the exponents δ and γ describing the damping time and the density of islands are related to each other as $\delta/\gamma \approx 4/3$ for the Wolf-Villain model and $\delta/\gamma \approx 2$ for the Family model. This result is in excellent agreement with recent theoretical predictions.

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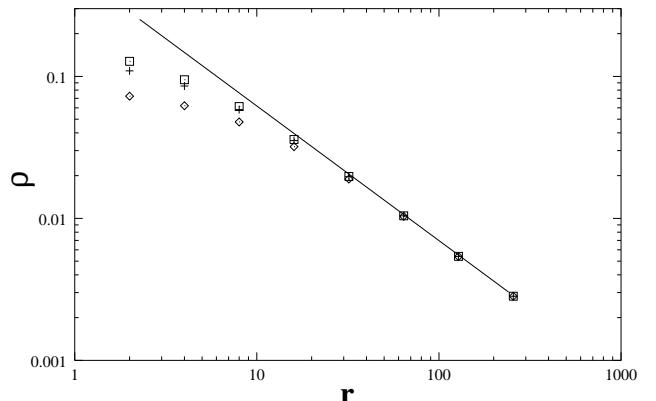


FIG. 1. Double logarithmic plot of density of islands ρ versus diffusion length r for the Wolf-Villain model in one dimension. The data are obtained from system size $L = 1000$ at different coverages, 0.1, 0.2, and 0.3 from the bottom, and are averaged over 1000 runs. The solid line with slope -0.94 is the guidance to the eyes.

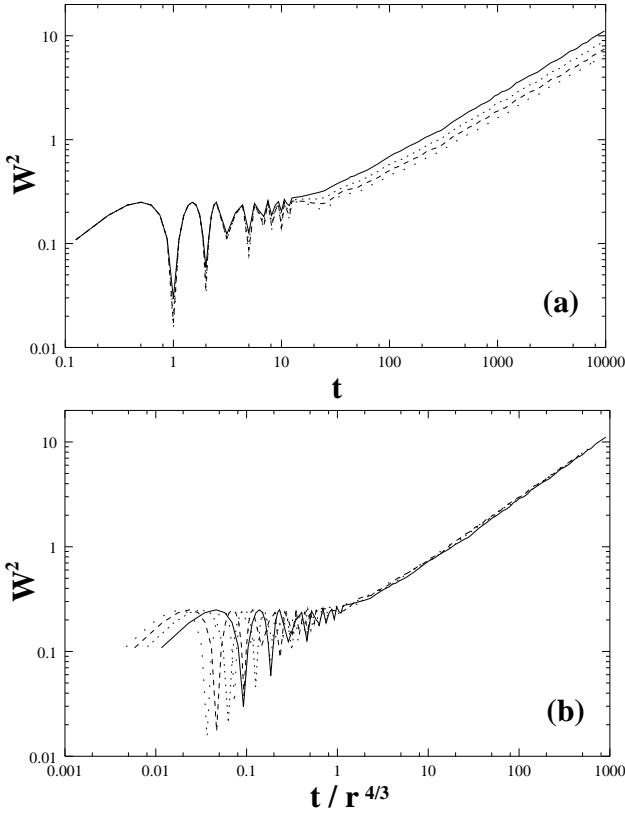


FIG. 2. Double logarithmic plot of the surface fluctuation width versus time t for the case (a) and rescaled time $t/r^{4/3}$ for the case (b) in the Wolf-Villain model. The data are obtained from system size $L = 1000$, and are averaged over 100 runs. The diffusion lengths $r = 6, 8, 10$ and 12 were used from the top.

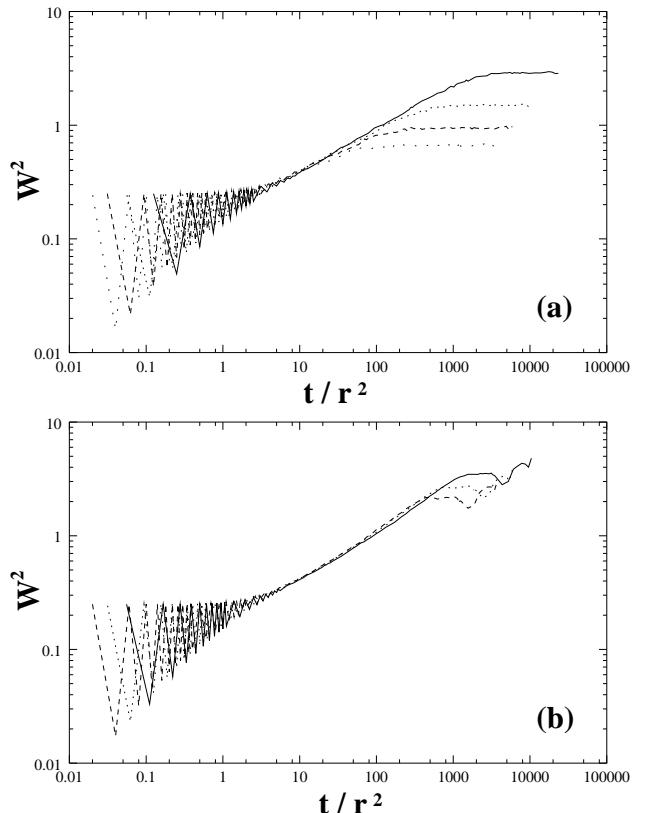


FIG. 3. Double logarithmic plot of the surface fluctuation width W^2 versus rescaled time t/r^2 in the Family model. In simulations, adatoms are allowed to run over humps for the case (a) and are not allowed to do for the case (b). For the case (a), various diffusion lengths, $r = 2, 3, 4$ and 5 are used, and for the case (b), $r = 3, 4$ and 5 are used. The data are obtained from system size $L = 1000$, and are averaged over 100 runs.